U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE ATTORNEY'S DOCKET NUMBER FORM PTO-139 (REV 10-05) TRANSMITTAL LETTER TO THE UNITED STATES P-001 ERM DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 INTERNATIONAL APPLICATION NO. INTERNATIONAL FILING DATE PRIORITY DATE CLAIMED PCT/RU98/00301 September 24, 1998 September 30, 1997 TITLE OF INVENTION X-Ray Absorbing Material and Variants APPLICANT(S) FOR DO/EO/US S) FOR DO/EO/US I.S. NOSOV; V. PETCHENKIN; and S.Y. SOKOLOV V.I. TKATCHENKO; V.A. IVANOV; Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: 1. X This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). 3. X A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. A copy of the International Application as filed (35 U.S.C. 371(c)(2)) is transmitted herewith (required only if not transmitted by the International Bureau). has been transmitted by the International Bureau. is not required, as the application was filed in the United States Receiving Office (RO/US). A translation of the International Application into English (35 U.S.C. 371(c)(2)). Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) are transmitted herewith (required only if not transmitted by the International Bureau). have been transmitted by the International Bureau. have not been made; however, the time limit for making such amendemnts has NOT expired. have not been made and will not be made. A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). Items 11. to 16. below concern document(s) or information included: An Information Disclosure Statement under 37 CFR 1.97 and 1.98. An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. A FIRST preliminary amendment. A SECOND or SUBSEQUENT preliminary amendment. A substitute specification. A change of power of attorney and/or address letter. 16. X Other items or information: (1) Declaration Claiming Small Entity Status(Independent Inventor) -five documents, one for each inventor (2) Conclusions of Preliminary Examination (3) International Search Report

| | norm se 17/CR TO P | ERNATIONAL APPLICATION NO | • | | ATTORNEY'S DOC | KET NUMBER |
|--|--|--|--|--|--|--------------------|
| U.S. APPLICATION NO (1) | 509256 | PCT/RU98/00301 | | 011 | P-001 E | |
| 17. X The foll | owing fees are submitted: | • | | CAL | CULATIONS | PTO USE ONLY |
| | AL FEE (37 CFR 1.492 (a) | | | | | |
| Search Repo | rt has been prepared by the I | EPO or JPO | \$880.00 | | | |
| International | preliminary examination fe | e paid to USPTO (37 CFR 1 | .482) \$680.00 | | | |
| | | n fee paid to USPTO (37 CFF O (37 CFR 1.445(a)(2)) | \$750.00 | | | |
| Neither interinternational | national preliminary examin search fee (37 CFR 1.445(a) | ation fee (37 CFR 1.482) not (2)) paid to USPTO | \$970.00 XXXXXXXXXXXXXXXXXXXXXXXXXXXXXXXXXX | | | |
| International and all claim | preliminary examination fees satisfied provisions of PC7 | e paid to USPTO (37 CFR 1. Article 33(2)-(4) | 482) \$94.00 | | | |
| | ENTER APPROP | RIATE BASIC FEE A | MOUNT = | s | 970.00 | |
| Surcharge of \$130 months from the e | 0.00 for furnishing the oath of arliest claimed priority date | or declaration later than (37 CFR 1.492(e)). | 20 30 | s | 0 | |
| CLAIMS | NUMBER FILED | NUMBER EXTRA | RATE | | | |
| Total claims | 5 -20 = | 0 | X \$22.00 | S | 0 | |
| Independent claims | 3 -3 = | 0 | X \$78.00 | \$ | 0 | |
| MULTIPLE DEPE | NDENT CLAIM(S) (if applica | | + \$250.00 | S | 0 | |
| | TOTAL C | F ABOVE CALCULA | TIONS = | \$ | 970.00 | <u> </u> |
| Reduction of 1/2 f must also by filed | for filing by small entity, if a (Note 37 CFR 1.9, 1.27, 1.2 | applicable. Verified Small E (8). | ntity Statement | S | 485.00 | |
| | | SUB | TOTAL = | \$ | 485.00 | |
| Processing fee of months from the e | \$130.00 for furnishing the E earliest claimed priority date | inglish translation later than (37 CFR 1.492(f)). | . 20 30 + | S | 0 | |
| | | TOTAL NATIO | NAL FEE = | S | 485.00 | |
| Fee for recording accompanied by a | the enclosed assignment (37 n appropriate cover sheet (3 | CFR 1.21(h)). The assignm 7 CFR 3.28, 3.31). \$40.00 p | ent must be er property + | S | 0 | |
| | | TOTAL FEES EN | CLOSED = | S | 485.00 | |
| | ************************************** | | | | ount to be: refunded | S |
| • | | | | | charged | 1.5 |
| | in the amount of $\frac{485}{}$ | to cover the ab | ove fees is enclose | a. | | |
| b. Please c A duplic | harge my Deposit Account 1 cate copy of this sheet is enc nmissioner is hereby authori ment to Deposit Account No | | fees which may beate copy of this sh | e requi | red, or credit | |
| b. Please of A duplic c. X The Corresponding NOTE: Where 1.137(a) or (b) | eate copy of this sheet is encommissioner is hereby authoriment to Deposit Account Notes an appropriate time limit must be filed and granted spondence to: | losed. ized to charge any additional | fees which may be cate copy of this shape of the shape of | e requi | red, or credit enclosed. | any |
| b. Please of A duplic c. The Coroverpay NOTE: Where 1.137(a) or (b) SEND ALL CORRES Lackenback One Chase Scarsdale, | eate copy of this sheet is encommissioner is hereby authoriment to Deposit Account Notes an appropriate time limit must be filed and granted spondence to: a Siegel Road b NY 10583 | losed. ized to charge any additional of the state of the | fees which may be cate copy of this shape of the shape of | net, a | red, or credit enclosed. | any |
| b. Please of A duplic c. The Coroverpay NOTE: Where 1.137(a) or (b) SEND ALL CORRES Lackenback One Chase Scarsdale, 914 723 43 Attorneys: | eate copy of this sheet is encommissioner is hereby authoriment to Deposit Account Notes an appropriate time limit must be filed and granted spondence to: a Siegel Road NY 10583 300 Henry A. Marzul | losed. ized to charge any additional of the charge any additional of the charge any additional of the charge and additional of the charge any additional of the charge and additional of the | fees which may be cate copy of this shape of the shape of | met, a | red, or credit enclosed. | any |
| b. Please of A duplic c. The Coroverpay NOTE: Where 1.137(a) or (b) SEND ALL CORRES Lackenback One Chase Scarsdale, 914 723 43 | eate copy of this sheet is encommissioner is hereby authoriment to Deposit Account Notes an appropriate time limit must be filed and granted spondence to: a Siegel Road NY 10583 | losed. ized to charge any additional of the charge any additional of the charge any additional of the charge and additional of the charge any additional of the charge and additional of the | fees which may be cate copy of this shape of the shape of | met, a pure la | med, or credit enclosed. petition to re Marzullo | any evive (37 CFR |

Serial or Patent No.: (to be assigned)

Page 1 of 2

P.09 14

Filed or Issued:

For: X-Ray Absorbing Material and Variants

DECLARATION CLAIMING SMALL ENTITY STATUS

[37 CFR 1.9(f) and 1.27(C)]

INDEPENDENT INVENTOR

As one of the five below named inventors,

| Inventor | Address |
|-------------------------------|---|
| Igor Stepanovitch NOSOV | Kv. 20, d. 43, ul. Sverdlova, Balashiha, Moskovskaya obl., 143900 RUSSIA |
| Vladimir Ivanovich TKATCHENKO | Kv. 20 d. 4, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Maleriy Anatolievich IVANOV | Kv. 29, d. 13/15, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Ivanovich PETCHENKIN | Kv. 181,d. 80, shosse Zaporozchsky, Dnepropetrovsk, 320041 THE UKRAINE |
| Stanislav Yurivich SOKOLOV | Kv. 62, d. 47, ul. Yuglas, Riga 1007 LATVIA |

I hereby declare that I qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under section 41(a) of Title 35, United States Code, to the Patent and Trademark Office with regard to the invention entitled X-Ray Absorbing Material and Variants described in

| \boxtimes | the specification | on filed herewith | |
|-------------|-------------------|-------------------|------------|
| | application ser | rial no. | , filed on |
| | Patent No. | , issued on | |

I have not assigned, granted, conveyed or licensed and am under no obligation under contract or law to assign, grant, convey or license, any rights in the invention to any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, or to any concern which would not qualify as a small business concern under 37 CFR 19(d) or a nonprofit organization under 37 CFR 1.9(e).

Each person, concern or organization to which I have assigned, granted, conveyed, or licensed or am under an obligation under contract or law to assign, grant, convey, or license any rights in the invention is listed below:

| X | no such person, concern, or organization |
|---|--|
| | persons, concerns, or organizations listed below |

| New National Phase Pul Application in the water of the part of 2 . State of 2 . | | |
|--|---|--|
| Note: Separate verified statements are requerganization having rights to the invention a CFR 1.27) | | - |
| FULL NAME: ADDRESS: | ☐ INDIVID <u>ÚAL</u> ☐ SMALL BUSINESS CONC ☐ NONPROFIT ORGANIZA | |
| FULL NAME: ADDRESS: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC ☐ NONPROFIT ORGANIZA | CERN |
| FULL NAME: ADDRESS: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC ☐ NONPROFIT ORGANIZA | |
| change in status resulting in loss of entitlem at the time of paying, the earliest of the issurdate on which status as a small entity is no large in that all statements made that all statements made on information and that these statements were made with the kethe like so made are punishable by fine or in Title 18 of the United States Code, and that statements of the validity of the application, any patent is verified declaration is directed. NAME OF INVENTOR | ent to small entity status e fee or any maintenand onger appropriate. (37 C de herein of my own know d belief are believed to la mowledge that willful fa aprisonment, or both, un uch willful false stateme | s prior to paying, or ce fee due after the CFR 1.28(b)) wledge are true and be true; and further alse statements and ader Section 1001 of ents may jeopardize |
| Stanislav Yurivich SOKOLOV | | |
| SIGNATURE OF INVENTOR | | |
| V (figure | | |
| DATE | | |

LACKENBACH SIEGEL MARZULLO ARONSON & GREENSPAN, P.C. SE-B-1

02.25.2000

-23-2000 09:32 Applicant or Patentee: Nosov et al.

Serial or Patent No.: (to be assigned)

Filed or Issued:

For: X-Ray Absorbing Material and Variants

914 723 4301 Docket No.: P-0001EK Page 1 of 2

DECLARATION CLAIMING SMALL ENTITY STATUS

[37 CFR 1.9(f) and 1.27(C)]

INDEPENDENT INVENTOR

As one of the five below named inventors,

| Inventor | Address |
|-------------------------------|---|
| Igor Stepanovitch NOSOV | Kv. 20, d. 43, ul. Sverdlova, Balashiha, Moskovskaya obl., 143900 RUSSIA |
| Vladimir Ivanovich TKATCHENKO | Kv. 20 d. 4, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Anatolievich IVANOV | Kv. 29, d. 13/15, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Ivanovich PETCHENKIN | Kv. 181,d. 80, shosse Zaporozchsky, Dnepropetrovsk, 320041 THE UKRAINE |
| Stanislav Yurivich SOKOLOV | Kv. 62, d. 47, ul. Yuglas, Riga 1007 LATVIA |

I hereby declare that I qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under section 41(a) of Title 35, United States Code, to the Patent and Trademark Office with regard to the invention entitled X-Ray Absorbing Material and Variants described in

| × | the specificati | on filed herewith | |
|---|-----------------|-------------------|------------|
| | application se | rial no. | , filed on |
| | Patent No. | , issued on | |

I have not assigned, granted, conveyed or licensed and am under no obligation under contract or law to assign, grant, convey or license, any rights in the invention to any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, or to any concern which would not qualify as a small business concern under 37 CFR 19(d) or a nonprofit organization under 37 CFR 1.9(e).

Each person, concern or organization to which I have assigned, granted, conveyed, or licensed or am under an obligation under contract or law to assign, grant, convey, or license any rights in the invention is listed below:

| X 1 | 10 | such | person, | concern, | or | organization |
|-----|----|------|---------|----------|----|--------------|
|-----|----|------|---------|----------|----|--------------|

persons, concerns, or organizations listed below.*

New Mational Phase PCT Application in the U.S. for "X-Kay Absorbing Material and Variants," Page 2 of 2

Note: Separate verified statements are required from each named person, concern or organization having rights to the invention evening to their status as small entities. (37 CFR 1.27)

| CFR 1.27) | | |
|---|---|------|
| FULL NAME: ADDRESS: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC ☐ NONPROFIT ORGANIZA | |
| FULL NAME: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC | |
| ADDRESS: | ☐ NONPROFIT ORGANIZA | TION |
| FULL NAME: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC | ERN |
| ADDRESS: | ☐ NONPROFIT ORGANIZA | TION |
| change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b)) I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 or lifte 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified declaration is directed. NAME OF INVENTOR | | |
| Valeriy Anatolievich IVANOV | | |
| SIGNATURE OF INVENTOR | | |
| Kubuutz | | |
| DATE ` | | |
| 02.25.2000 | | |

LACKENBACH SIEGEL MARZULLO ARONSON & GREENSPAN, P.C. SE-B-1

DOCKELING., F.

Page 1 of 2

P.09 14

Applicant or Patentee: Nosov et al.

Serial or Patent No.: (to be assigned)

Filed or Issued:

For: X-Ray Absorbing Material and Variants

DECLARATION CLAIMING SMALL ENTITY STATUS

[37 CFR 1.9(f) and 1.27(C)]

INDEPENDENT INVENTOR

As one of the five below named inventors,

| Inventor | Address |
|-------------------------------|---|
| Igor Stepanovitch NOSOV | Kv. 20, d. 43, ul. Sverdlova, Balashiha, Moskovskaya obl., 143900 RUSSIA |
| Vladímir Ivanovich TKATCHENKO | Kv. 20 d. 4, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Anatolievich IVANOV | Kv. 29, d. 13/15, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Ivanovich PETCHENKIN | Kv. 181,d. 80, shosse Zaporozchsky, Dnepropetrovsk, 320041 THE UKRAINE |
| Stanislav Yurivich SOKOLOV | Kv. 62, d. 47, ul. Yuglas, Riga 1007 LATVIA |

I hereby declare that I qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under section 41(a) of Title 35, United States Code, to the Patent and Trademark Office with regard to the invention entitled X-Ray Absorbing Material and Variants described in

| \boxtimes | the specification | on filed herewith | |
|-------------|-------------------|-------------------|------------|
| | application ser | rial no. | , filed on |
| | Patent No. | , issued on | |

I have not assigned, granted, conveyed or licensed and am under no obligation under contract or law to assign, grant, convey or license, any rights in the invention to any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, or to any concern which would not qualify as a small business concern under 37 CFR 19(d) or a nonprofit organization under 37 CFR 1.9(e).

Each person, concern or organization to which I have assigned, granted, conveyed, or licensed or am under an obligation under contract or law to assign, grant, convey, or license any rights in the invention is listed below:

| X | no such person, co | ncern, or organization |
|---|--------------------|---------------------------------|
| | persons, concerns, | or organizations listed below.* |

FEB-23-2000 09:35 LACKENBACH STEGEL 914 723 4301 P.08 14 New National Phase PUT Application in the U.S. for "A-kay absorbing manerial and Variants," Page 2 of 2

Note: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities. (37 CFR 1.27)

| OT IT AID. | | | |
|--|--|-----|--|
| FULL NAME: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC | ERN | |
| ADDRESS: | □ NONPROFIT ORGANIZA | | |
| FULL NAME: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC | FRN | |
| ADDRESS: | ☐ NONPROFIT ORGANIZA | | |
| FULL NAME: | ☐ INDIVIDUAL ☐ SMALL BUSINESS CONC | ERN | |
| ADDRESS: | ☐ NONPROFIT ORGANIZA | | |
| I acknowledge the duty to file, in this application or patent, notification of an change in status resulting in loss of entitlement to small entity status prior to paying, of at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b)) I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which the verified declaration is directed. | | | |
| Valeriy Ivanovich PETCHENKIN | NAME OF INVENTOR Valeriy Ivanovich PETCHENKIN | | |
| | | | |
| SIGNATURE OF INVENTOR | | | |
| Breed | | | |
| DATE | | | |
| 01 15 1000 | | | |

LACKENBACH SIEGEL MARZULLO ARONSON & GREENSPAN, P.C. SE-B-1

Applicant or Patentee: Nosov, et al.

etal.

DOCKELINO.. F-0001EA

P.09 14

Page 1 of 2

Serial of Patent No.: (to be assigned) Filed or Issued:

For: X-Ray Absorbing Material and Variants

DECLARATION CLAIMING SMALL ENTITY STATUS

[37 CFR 1.9(f) and 1.27(C)]

INDEPENDENT INVENTOR

As one of the five below named inventors,

| Inventor | Address |
|-------------------------------|---|
| Igor Stepanovitch NOSOV | Kv. 20, d. 43, ul. Sverdlova, Balashiha, Moskovskaya obl., 143900 RUSSIA |
| Vladimir Ivanovich TKATCHENKO | Kv. 20 d. 4, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Anatolievich IVANOV | Kv. 29, d. 13/15, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Ivanovich PETCHENKIN | Kv. 181,d. 80, shosse Zaporozchsky, Dnepropetrovsk, 320041 THE UKRAINE |
| Stanislav Yurivich SOKOLOV | Kv. 62, d. 47, ul. Yuglas, Riga 1007 LATVIA |

I hereby declare that I qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under section 41(a) of Title 35, United States Code, to the Patent and Trademark Office with regard to the invention entitled X-Ray Absorbing Material and Variants described in

| \boxtimes | the specification | on filed herewith | |
|-------------|-------------------|-------------------|------------|
| | application se | | , filed on |
| | Patent No. | , issued on | , |

I have not assigned, granted, conveyed or licensed and am under no obligation under contract or law to assign, grant, convey or license, any rights in the invention to any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, or to any concern which would not qualify as a small business concern under 37 CFR 19(d) or a nonprofit organization under 37 CFR 1.9(e).

Each person, concern or organization to which I have assigned, granted, conveyed, or licensed or am under an obligation under contract or law to assign, grant, convey, or license any rights in the invention is listed below:

| X | no such person, concern, or organization |
|---|--|
| | persons, concerns, or organizations listed below.* |

FEB-23-2000 09:35 LACKENBACH SIEGEL 914 723 4301 P. 08 14 New National Phase FC1 Application in the U.S. for "Arkey Australia material and Variants," Page 2 of 2

Note: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities. (37 CFR 1.27) FULL NAME: ☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN □ NONPROFIT ORGANIZATION ADDRESS: □ INDIVIDUAL **FULL NAME:** ☐ SMALL BUSINESS CONCERN □ NONPROFIT ORGANIZATION ADDRESS: **FULL NAME:** ☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN ADDRESS: ☐ NONPROFIT ORGANIZATION I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b)) ill hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 8 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified declaration is directed. NAME OF INVENTOR Vladimir Ivanovich TKATCHENKO SIGNATURE OF INVENTOR DATE 02.25,2000

LACKENBACH SIEGEL MARZULLO ARONSON & GREENSPAN, P.C. SE-B-1

914 723 4301 DOCKELNO.: r-vuvien

P.07 14

Applicant or Patentee: Nosov et al.

Serial or Patent No.: (to be assigned)

Filed or Issued:

For: X-Ray Absorbing Material and Variants

Page 1 of 2

DECLARATION CLAIMING SMALL ENTITY STATUS

[37 CFR 1.9(f) and 1.27(C)]

INDEPENDENT INVENTOR

As one of the five below named inventors,

| Inventor | Address |
|-------------------------------|---|
| Igor Stepanovitch NOSOV | Kv. 20, d. 43, ul. Sverdlova, Balashiha, Moskovskaya obl., 143900 RUSSIA |
| Vladimir Ivanovich TKATCHENKO | Kv. 20 d. 4, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Anatolievich IVANOV | Kv. 29, d. 13/15, pr. K. Marksa, Dnepropetrovsk, 320095 THE UKRAINE |
| Valeriy Ivanovich PETCHENKIN | Kv. 181,d. 80, shosse Zaporozchsky, Dnepropetrovsk, 320041 THE UKRAINE |
| Stanislav Yurivich SOKOLOV | Kv. 62, d. 47, ul. Yuglas, Riga 1007 LATVIA |

I hereby declare that I qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under section 41(a) of Title 35, United States Code, to the Patent and Trademark Office with regard to the invention entitled X-Ray Absorbing Material and Variants described in

| \boxtimes | the specification | on filed herewith | |
|-------------|-------------------|-------------------|------------|
| | application ser | rial no. | , filed on |
| | Patent No. | , issued on | |

I have not assigned, granted, conveyed or licensed and am under no obligation under contract or law to assign, grant, convey or license, any rights in the invention to any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, or to any concern which would not qualify as a small business concern under 37 CFR 19(d) or a nonprofit organization under 37 CFR 1.9(e).

Each person, concern or organization to which I have assigned, granted, conveyed, or licensed or am under an obligation under contract or law to assign, grant, convey, or license any rights in the invention is listed below:

| X | no such | person, | concern, | or | organization |
|---|---------|---------|----------|----|--------------|
|---|---------|---------|----------|----|--------------|

| | • | • | ~ | | |
|----------|-----------|-------------|-------|--------|---------|
| persons, | concerns, | or organiza | tions | listed | below.* |

FEB-23-2000 09:33 LACKEMBACH STEGEL 914 723 4301 P.06 14 NEW NATIONAL PROSERVE APPLICATION IN THE U.S. FOR "A-RAY ADSORDING MATERIAL AND Variants," Page 2 of 2 Note: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities. (37 CFR 1.27) **FULL NAME: INDIVIDUAL** ☐ SMALL BUSINESS CONCERN □ NONPROFIT ORGANIZATION ADDRESS: □ INDIVIDUAL **FULL NAME:** ☐ SMALL BUSINESS CONCERN □ NONPROFIT ORGANIZATION ADDRESS: **FULL NAME:** ☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN ADDRESS: □ NONPROFIT ORGANIZATION I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b)) I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the application, any patent issuing thereon, or any patent to which this verified declaration is directed. NAME OF INVENTOR Igor Stepanovitch NOSOV SIGNATURE OF INVENTOR Mcekel) DATE

LACKENBACH SIEGEL MARZULLO ARONSON & GREENSPAN, P.C. SE-B-1

02.25,2000

09/509256 416 Rec'd PCT/PTO 2 2 MAR 2000

X-RAY ABSORBING MATERIAL (VARIANTS).

Field of engineering.

The invention relates to X-ray contrasting and X-ray protection materials and can be used in medicine, namely in roentgen equipment intended for diagnostics and inspection ill, specifically for monitoring over condition of endo-prosthetic appliances, internal surgical wields, post-surgical area in order to avoid leaving a surgical napkin, tampon or surgical instruments inside the body of a patient, for selection of exposure areas in the course of radio-therapy, etc., as well as for production of protective uniform (aprons, smocks, waistcoats, caps, etc.), production of protection shields, partitions, protection coating, isolation materials, etc.

Previous level of engineering.

Known is an X-ray absorbing material, for example, under the Patent of Sweden № 349366, 1960, providing for an artificial rayon thread which contains barium sulfate (BaSO4) in the form of mechanical impurity (15 up to 65 % of mass). However, adding of the mentioned mechanical impurity to the textile base of material results in abrupt reduction of its durability.

Known are X-ray absorbing materials, executed, for example, in the form of threads which contain bismuth oxide, colloidal silver, iodine derivatives as X-ray contrasting impurities added to the polymeric composition (ref. to X-ray absorbing materials described, for example, in the Abstract of A. V. Vitulsky, Master of Science, named «Obtaining and research of synthetic fibers with the X-ray contrasting and anti-germ preparations being added at the time of forming», Leningrad, 1974).

However, examining of properties of a textile base containing such impurities has shown that due to violation of the fiber structure homogeneity, caused by negative influence of contrasting impurity particles, the worsening of the physical and mechanical properties of fibers and threads made on the base of the mentioned impurities takes place. A textile base containing such impurities lacks durability and this factor limits the field of application thereof.

Known is an X-ray absorbing material, for example, under the Certificate of Authorship of Bulgaria № 36217, 1980, made in the form of a thread containing X-ray protection coating produced of heavy metals, plotted by means of falling out in corresponding salts solutes. Unlike the materials mentioned above, this one has better physical and mechanical properties

since the plotting of the coating by falling out of heavy metals from solute does not really affect the mechanical properties of the initial material. Nevertheless, the small width of the coating causes the lowered X-ray contrasting and X-ray protection properties. Furthermore, the weak adhesion of the X-ray absorbing coating towards initial material after washing, cleaning and so on, causes abrupt reduction of X-ray contrasting and X-ray protective properties.

Known is an X-ray absorbing material under the Certificate of Authorship No 1826173 A616 17/56, 17/00, U.S.S.R., 1980, which, having the merits of a material made in the form of the thread containing the X-ray absorbing coating of heavy metals, is devoid of its drawbacks due to the fact that the X-ray absorbing coating is made of ultra dispersible particles (UDP) with sizes of between 10^{-6} and 10^{-7} m and having such property like abnormal weakening of radiation (according to «Phenomenon of abnormal reduction of X-ray radiation by ultra dispersible environment», Diploma No 4 of the Russian Natural Science Academy, priority date - 05/07/87). The metal-containing element (size of between -10^{-6} and 10^{-7} m) finely dispersible mixture of this material is bonded to the surface of thread *i. e.* on the textile base surface. However, the use of finely dispersible mixture only in the range of ultra dispersible particles (between 10^{-6} and 10^{-7} m), which are chemically and physically fissile and pyrophoric, is technologically problematic since it requires special conditions of manufacture, transporting, storage and technological application.

As a result of the recent discovery in the field of physics of poly-dispersed environment named «The phenomenon of the permeating radiation quantum stream intensity abnormal alteration by mono- and multiple environment» (Diploma № of the Russian Natural Science Academy, priority date - 09/19/96) it was ascertained that the poly-dispersed environment, provided that the certain dispersibility of particles and segregation thereof by intermixing is ensured, also reveals the capability of abnormally high reduction of X-radiation, which is conditioned by self-organization of poly-dispersed particles having a size of between one thousandth and hundreds of micrometers into energetically interconnected X-ray absorbing ensembles. (Segregation of poly-dispersed mixture denotes irregular distribution of the poly-dispersed mixture particles caused by intermixing of the mixture, due to self-organization of particles into the system of energetically interconnected ensembles ensuring the increasing of the photo-absorption cut). Meanwhile it is generally known that the use of poly-dispersed mixtures consisting of particles having a size of between 10-9 up to 10-3 in modern engineering does not require any specific limitations and is not fraught with any specific technological difficulties in manufacture, transportation, storage and use.

Known is an X-ray absorbing material containing, for example, a rubber matrix with a fixed X-ray absorbing filler under the U. S. patent № 3239669, 1966. According to the patent, the X-ray absorbing elements in the form of lead, bismuth, silver and tungsten can be used as a filler. The main drawback of the mentioned material is reduction of solidity of material in 2-3 times due to the negative influence of the absorbing particles of filler violating the uniform structure of the original polymeric mass.

Known are another X-ray absorbing materials containing a matrix with a fixed X-ray absorbing filler or, for example, in the form of golden tubes, under the U. S. patent № 2153889, 1939 or in the form of wire made of silver-, bismuth-, tantalum-containing alloys, wherein the said wire and the matrix are fastened together by interweaving and forming a kind of a textile thread (U. S. patent № 3194239, 1965).

Materials containing a matrix with a fixed X-ray absorbing filler in the form of wire made of silver-, bismuth-, tantalum-containing alloys, wherein the said wire and the matrix are fastened together by interweaving and are forming a textile thread, are preferable in comparison with the materials under the U.S. patent № 2153889, if taking into account such property as solidity, but have lower plasticity, which is inadmissible in many cases.

Known are materials protecting from impact of X- and gamma-radiation containing heavy fillers, the most spread of which is, for example, lead (Article named «Technical headway in atomic engineering», Series «Isotopes in U. S. S. R», 1987, edition 1(72), p. 85). Due to the great difference in density between a filler (for example, lead) and a matrix (for example, concrete, polymers, etc.) the filler (lead) is being spread along the matrix volume irregularly which results in decrease of the X-ray absorbing properties of material as a whole.

Known is an X-ray absorbing material executed, for example, on the basis of the polysterol polymeric matrix and lead-containing organic filler, under the U.K. patent № 1260342, G 21 F 1/10, 1972. The said material has the same drawback as the lead-containing fillers described in the article «Technical headway in atomic engineering», Series «Isotopes in U. S. S. R», 1987, edition 1(72), p. 85, which consists in irregular distribution of heavy X-ray absorbing filler inside the matrix, the material of which has considerably lower density than the material of filler.

The closest to the offered invention is an X-ray absorbing material containing a matrix with the fixed X-ray absorbing metal-containing filler in the form of dispersed particles, under the Russian Federation patent № 2063074 G21 F 1/10 of 06/27/96 (prototype). The drawback of the said material consists in the fact that adding of a lead-containing filler to a textile base results in reduction of density of the material due to the violation of the textile base uniform structure, that limits, in its turn, the possibility of use thereof for manufacture of various

protective means. Material executed on the basis of a thread with a lead-containing filler can not be used as an X-ray contrasting material in medical radiology due to the lead's toxic properties. Furthermore, it is impossible to create effective compact protection from X- and gamma-radiation on the basis of such material as a thread (analogue thereof, for example, is described in the Russian Federation patent № 2063074) as in this case for purposes of using the said thread-material it is necessary to apply the special technology of dense multi-layer machine knitting for manufacture of multipurpose protective tissue. But thus, as the weakening of a narrow bundle of quantums by a stratum of a material having width = X happens according to the exponential law, in compliance with the legitimacy described in the book "Methods of radiation granulometry and statistical simulation in research of structural properties of composite materials. " (V. A. Vorobiev, B. E. Golovanov, S. I. Vorobieva, Moscow, Energoatomizdat, 1984), happens reduction of radiation intensity:

$$I=Io e^{-\mu x}$$
 (1)

Where

I is the intensity of radiation passed through a stratum of material having width = X, lo is the intensity of initial radiation,

μ is the linear factor of radiation reduction (weakening) (tabular regulated value for each of X-ray absorbing materials).

The drawback of the prototype consists also in high percentage of a metal-containing filler in total amount of the X-ray absorbing material (66 - 89 %), that will cause increase of the mass of X-ray absorbing material as a whole, and on the other hand, the articles made out of such material are heavy and inconvenient in maintenance.

The irregular distribution of the heavy filler in the matrix volume is one more drawback of the mentioned prototype.

Disclosure of invention

The main tasks in the course of development of X-ray absorbing (i. e. X-ray contrasting and X-ray protective) materials are:

- to eliminate toxicability of an X-ray contrasting material,
- to reduce the mass and width of a protective material.

Elimination of toxicability is achieved through the application of non-toxic fillers (tungsten, for example). And creation of compact protection with the protective material width reduced together with saving of X-ray absorbing properties (i. e. X-ray and gamma-radiation reduction degree) leads to increasing of the material protective layer mass caused by use of "heavy" fillers, i. e. filler of high density. Vice versa, when the X-ray absorbing properties are saved, reduction of protective material density causes the necessity of increasing of its width.

Let's illustrate this position on an example of an X-ray absorbing material in the form of a protective tissue (radiologist' protective apron, for example) which ensures protection characterized by the reduction factor K=100. It is possible to deduce from Formula (1) as follows:

$$K = I_0/I = e^{\mu x} = 100$$
,

Whence it follows

$$x=\ln K/\mu=4,6/\mu$$
 (2)

As an example, let's compare properties of tissues made of threads containing the known fillers in the form of non-segregated dispersed particles of lead (Pb) and tungsten (W). The size of the compared tissues was set as 10×10 cm. The rest initial data for comparison are shown in Table 1.

Table 1.

Initial data for comparison.

| Material used for the particles of | Linear | factor | of | radiation | Particles material | density, |
|------------------------------------|-----------|------------|-----|-----------|--------------------|----------|
| filler | reduction | n | | | ρg/sm ³ | |
| | | -1 | | | | |
| | (weaken | ing), μ, c | m*) | | | |
| Pb | 40,3 | | | | 11,34 | |
| W | 50,1 | _ + -,+ ,+ | | | 18,7 | |

^{*)} Remark: radiation source is an X-ray emitting (roentgen) tube, energy – 60 keV.

It is possible to deduce from Formula (2), using the data of Table 1, the values of width X for tissues made of threads with a filler made of:

Pb (X = 0.11cm) and of W (X = 0.09 cm).

Accordingly the mass of such protection tissues with volume of 10 x 10 x X will constitute:

for
$$W - 168,3$$
 g.

If the mass of a protection tissue on the Pb basis is taken for 1, then (by equal protective properties and equal sizes) the ratio of masses of tissues made on the base of threads containing Pb and W - will be 1:1,35.

Thus, it is impossible to obtain simultaneous reduction of the protective material width and mass using the prototype and known similar technologies.

According to the present invention the set tasks are solved by means mentioned in the distinctive part of independent claims of the invention formula.

In a first embodiment of an X-ray absorbing material comprising a matrix with fixed X-ray absorbing metal-containing filler, the said material uses as a filler the segregated by intermixing poly-dispersed mixture containing metallic particles having a size of between 10⁻⁹ and 10⁻³, while the textile base serves as a matrix. As this takes place, the particles are bonded to the surface of said textile base and the density of X-ray absorbing material as a whole, at X-ray absorbing properties of the material being equal to those of the material used for the particles of the X-ray absorbing filler, is defined by the relation:

$$\rho m = (0.01 - 0.20)\rho_p$$

where: ρ m - density of X-ray absorbing material as a whole,

while ρ_P is the density of the material used for the particles of the X-ray absorbing filler.

In a second embodiment of an X-ray absorbing material comprising a matrix with fixed X-ray absorbing metal-containing filler in the form of dispersed particles, the said material uses as a filler the segregated by intermixing poly-dispersed mixture containing metallic particles having a size of between 10⁻⁹ and 10⁻³ m, wherein the said metallic particles are surrounded by the volume of a matrix executed of at least one component solidifying under atmospheric pressure or of the composition on the base of said component. As this takes place

the total mass of segregated poly-dispersed mixture consisting of X-ray absorbing particles of filler, is defined by the relation:

$$M = (0.05 - 0.5) \text{ m}$$

where M is the total mass of segregated poly-dispersed mixture consisting of X-ray absorbing particles of filler,

while m is the equivalent mass of the X-ray-absorbing filler material equal by its protective properties to the mass M.

In a third embodiment of an X-ray absorbing material comprising a matrix with fixed X-ray absorbing metal-containing filler in the form of dispersed particles, the said material uses as a filler the segregated by intermixing poly-dispersed mixture, containing metal particles having a size of between 10⁻⁹ up to 10⁻³ m, wherein the said particles are bonded to an intermediate substrate which is surrounded by the volume of matrix executed of at least one compound solidifying under atmospheric pressure or of the composition on the base of said compound. A textile base serves as an intermediate substrate. A mineral fiber can be used as an intermediate substrate.

The attributes set forth above relate to a range of inventions interconnected by the common author's conception. As this takes place, the said range of inventions consists of objects of one type and appliance, ensuring the same technical result, namely: exclusion of toxicability of an X-ray contrasting material and reduction of mass and width of a protective material which is the necessary requirement for an invention represented by variants.

Inventions realization variants.

In a first embodiment of X-ray absorbing material the execution of a filler in the form of segregated by intermixing poly-dispersed mixture comprising metallic particles having a size of between 10⁻⁹ up to 10⁻³ m ensures the manifestation of a qualitatively new effect by used X-ray absorbing filler - increasing of cut of interaction between the X-ray and gamma-ray emission and substance. Due to the mentioned effect the increasing of specific properties of X-ray absorption with the offered material is achieved.

The use of poly-dispersed mixtures as a filler is widely applied in X-ray absorbing materials described, for example, in the Russian Federation patents № 2063074 and №

2029399, whereof the non-segregated particles having a size of between 10⁻⁶ up to 10⁻³ m are used. However, in the said materials the above mentioned attribute is used in purpose of more regular distribution of X-ray absorbing filler along the surface of a matrix or inside thereof.

In the X-ray absorbing metal-containing material defined herein the segregated by intermixing poly-dispersed mixture ensures not only the more regular distribution of X-ray absorbing filler along the surface of a matrix or inside thereof but also provides for manifestation of a qualitatively new effect - increasing of cut of interaction between the X-ray and gamma-ray emission and substance.

The finely dispersible mixture of metal-containing element (sizes between 10⁻⁶ and 10⁻⁷ m) used in the known analogous material under the U.S.S.R. Certificate of Authorship № 1826173 is bonded to the textile base surface. Unlike the said analogous material, under the offered invention the poly-dispersed mixture made of particles having sizes of wide range: 10⁻⁹ up to 10⁻³, is used. As this takes place, the particles having sizes of the above mentioned range are included into the common mixture. Consequently, working on such mixture in common, natural conditions does not reveal any technological obstacles, *i.e.* the said mixture does not demonstrate physical and chemical activity. In particular it does not manifest pyrophoric properties.

Under the offered invention the use of the segregated by intermixing poly-dispersed mixture comprising particles having sizes in the range of 10⁻⁹ up to 10⁻³ m provides for a qualitatively new effect, if compared with the said analogous material under the U.S.S.R. Certificate of Authorship № 1826173. This effect consists in obtaining the same abnormal X-ray absorbing properties.

Side by side with this, the dispersed particles of the analogous material (acc. to the U.S.S.R. Certificate of Authorship № 1826173) are bonded to the thread surface, *i.e.* to the surface of a textile base. However, under the offered invention not only a thread but also separate filaments thereof can be used as a textile base, *i.e.* the notion «textile base» grasps a thread as well as separate filaments. According to the present invention in the case of separate filaments coating by an X-ray-absorbing filler (and, what is more, in the form of segregated by intermixing poly-dispersed mixture with self-organization of poly-dispersed particles into the energetically interconnected power-consuming ensembles), and provided that the filaments would twist into a thread, the said thread shall have the specific X-ray absorbing properties of a qualitatively new, higher level, in comparison with the analogous material under the U.S.S.R. Certificate of Authorship № 1826173.

So, using a textile base as a matrix with the X-ray absorbing metal-containing segregated particles of filler being bonded to surface thereof ensures a qualitatively new effect

(differing from the prototype) which finds expression in higher X-ray absorbing properties of material characterized by extremely heightened specific properties of X-ray absorption.

Under the U.S.S.R. Certificate of Authorship № 1826173 an X-ray absorbing coating of the thread-matrix surface is provided. As for the X-ray absorbing material offered herein, a textile base not only in the form of thread as a whole can be used as a matrix, but also a textile base in the form of separate filaments which the thread consists of (as mentioned above). A thread twisted out of separate filaments coated by an X-ray absorbing filler has much higher X-ray absorbing properties than a thread where only the open surface thereof is coated with an X-ray absorbing filler (unlike the offered material, where the surface of each filament included into the thread is coated by an X-ray absorbing filler). Moreover, the surface of each filament is covered by dispersed particles segregated by intermixing. As a result the said dispersed particles are self-organized into the energetically interconnected X-ray absorbing ensembles, and this, in its turn, ensures the extreme heightening of X-ray absorbing specific characteristics.

Realization of an X-ray absorbing material as a whole, at same X-ray absorbing properties of this material and the filler material, wherein the density of filler is defined by the relation:

$$\rho_{m} = (0.01 - 0.20)\rho_{p}$$

where ρ m is density of X-ray absorbing material as a whole,

while ρ_P is the density of the material used for the particles of the X-ray absorbing filler,

creates a qualitatively new effect (if compared with the material of the prototype), namely the simultaneous reduction of width and density of a protective material.

The simultaneous reduction of width and density of a protective material woven, for example, of an X-ray absorbing thread, ensures overcoming of the main contradiction arising while creating effective compact protection against X- and gamma-radiation. According to the offered invention, densities of the protective materials in the form of a thread and tissues derived therefrom, depending on the set technical conditions, can constitute between 0,01 (upper limit) and 0,2 (lower limit) of the X-ray absorbing filler particles material density. If the mass of X-ray absorbing material (in the present case, a protective tissue executed on the base of a thread, according to the invention) is taken for 1, then at protective properties and

Table 2

sizes of compared protective tissues being equal to those of the tissue on the basis of the offered thread, for conditions set forth in Table 1, the correlation by mass will be as defined in Table 2 below.

Comparative correlation of tissues by masses at equal protection properties (with regard to the data set forth in Table 1)

| Relative limits of | Tissue made of | Tissue made of | Tissue made of |
|----------------------|------------------|--------------------|--------------------|
| oscillation of | offered material | threads with a | threads with a |
| correlation | | filler in the form | filler in the form |
| between density of | | of non-segregated | of non-segregated |
| tissue made of the | | particles of Pb | particles of W |
| offered material | | | |
| and density of the | | | |
| material used for | | | |
| the particles of the | | | |
| X-ray absorbing | | | |
| filler | | | |
| Upper limit (0,01) | 1 | 198 | 267 |
| Lower limit (0,2) | 1 | 9,9 | 13,35 |

So, the offered X-ray absorbing material (tissue) would have mass lesser in 9,9 up to 267 times (at the other physical and chemical parameters being equal), if compared with the protection tissues on the basis of threads with a filler in the form of non-segregated particles of Pb and W. The mentioned factor ensures a qualitatively new effect.

Consequently, if compared with the prototype, the offered X-ray absorbing material, demonstrating the absolute absence of toxicability, ensures high solidity equal to the solidity of the X-ray absorbing textile base plotted. Furthermore, it ensures the abnormally high X-ray absorbing properties at low density.

In a second embodiment of X-ray absorbing material the use of segregated by intermixing poly-dispersed mixture comprising metallic particles having a size of between 10⁻⁹ up to 10⁻³ m (as set forth above) ensures the manifestation of a qualitatively new effect by

the used X-ray absorbing filler - increasing of cut of interaction between the X-ray and gamma-ray emission and substance.

The poly-dispersed mixture containing metallic particles having a size of between 10⁻⁹ up to 10⁻³ m, being placed inside a matrix volume, wherein the matrix is made of at least one component solidifying under atmospheric pressure or of a composition on the basis of said component, excluded is violation of energetic X-ray absorbing ensembles formed by intermixing and made of the X-ray absorbing element particles segregated poly-dispersed mixture. Meanwhile, this promotes the self-organization of said energetic X-ray absorbing ensembles.

An inorganic glue, such as the Na silicate and K silicate water solute or water suspension of compositions containing oxides of alkaline metals and earth metals, as well as compositions on the base of such glue, can be used as a matrix.

The natural polymers, such as collagen, albumin, casein, gum, wood pitch, starch, dextrin, latex, natural caoutchouc, gutta-percha, zein, soy casein, as well as compositions on the base of such polymers, can also be used as a matrix.

The synthetic polymers, such as polyakrylates, polyamides, polyethylenes, polyethers, polyurethanes, synthetic rubber, phenolformaldehyde resins, carbomid resins, calibration epoxy and compositions based on such polymers, can be used as a matrix.

Element-organic polymers, such as silicon-organic polymers, boron-organic polymers, metal-organic polymers and compositions based on such polymers, can also be used as a matrix.

Plastics filled with gas, such as foam plastic and expanded plastic, can be used as a matrix.

Vegetable oils or drying oils can be used as a matrix.

Solutes of film-generating substances, such as oily, alkyd, ether-cellulose lacquers, can be used as a matrix.

Polymers water dispersions, such as emulsion colors, can be used as a matrix.

Concrete, gyps and so on can be used as a matrix.

According to the invention defined herein using the matrix made of solidifying compound, unlike the material-prototype under the Russian Federation patent № 2063074, takes place under atmospheric pressure, *i.e.* under natural conditions and not under pressure of 150 mPa like according to the prototype. According to the invention defined herein the mixture is not underwent pressure like the protection rubbers as described in the Russian Federation patents №№ 2077745, 2066491, 2069904 which are underwent vulcanization under pressure after

preparation of the mixture. Consequently, it helps to avoid destroying of energetic X-ray absorbing ensembles formed in the course of intermixing of X-ray absorbing element particles segregated poly-dispersed mixture. The same distinction of the invention defined herein from the analogous material under the U.S.S.R. Certificate of Authorship № 834772 takes place, since according to the mentioned Certificate an X-ray absorbing material is obtained under pressure of 150-200 kg/cm²

In an analogous material under the U.S. patent № 3194239 the pressed pills of previously crumbled up iron-manganese concretions (IMC) are used as an X-ray absorbing filler, which is different from the invention defined herein. Effect of pressure on the filler of an analogous material under the Russian Federation patent № 2029399 also results in impossibility of energetical ensembles self-organizing (however it takes place in the offered invention). Thus, application as a matrix of at least one compound solidifying under atmospheric pressure or of compositions on its base in the offered invention has essential differences from the material - prototype as defined in the Russian Federation patent № 2063074 7. and from the analogous materials under the Russian Federation patents №№ 2029399, 2077745, 2066491, 2069904 in part of respective functional properties.

Realization of a condition, at which the common mass of the segregated poly-dispersed mixture consisting of the X-ray absorbing filler particles material is defined by the relation

$$M = (0.05 - 0.5) \text{ m},$$

where M is a the total mass of segregated poly-dispersed mixture consisting of X-ray absorbing particles of filler;

while m is equivalent mass of the X-ray absorbing filler material which is equal by its protective properties to the mass M,

- will allow (according to the second variant of X-ray absorbing material) to reduce a mass of known X-ray absorbing fillers in protection materials in 2 up to 20 times, depending on particular technical conditions and at saving an X-ray and gamma-ray radiation reduction factor.

Reduction of mass and width of protection material can be regarded as the main objective while constructing protection from roentgen- and gamma-radiation. However creation of the compact protection having a diminished thickness of layer leads to increase of a protective layer mass because of usage of the known heavy fillers. And, vice versa, saving of a roentgen - and the gamma - radiation reduction factor at lowering the density of a material entails necessity of increasing width of protection. And this is the main inconsistency arising while creating effective compact protection from roentgen - and gamma-radiation, as

the simultaneous reduction of width and mass of X-ray absorbing material practically cannot be achieved with the known fillers applied for protection. This inconsistency requires some compromise approach as to the choice of protection width and mass with allowance for a cost of such protection.

Let's illustrate this problem on an example of the most common material applied in purpose of protection against gamma-radiation, such as concrete. Density of different sorts of usual Portland concrete, containing cement as a connecting substance and the silicon shingle, gravel, quartz sand and similar mineral fillers, constitutes 2,0 - 2,4 g / cm³. A linear gamma-radiation reduction factor constitutes 0,11 - 0,13 cm⁻¹ (for energies of 1-2 MeV). The protection made of concrete having such density is quite cumbrous and should have considerable width. The concrete containing cement as a connecting substance, sand as a filler and galena as an X-ray absorbing filler in the ratio 1: 2: 4 has the density of 4,27 g/cm³, and the linear reduction factor thereof constitutes 0,26 cm⁻¹ (for energies 1,25 MeV). The concrete containing cement as a connecting substance, sand as a filler and lead as an X-ray absorbing filler in the ratio 1: 2: 4 has density of 5,9 g/cm³, and the linear reduction factor thereof constitutes 0,38 cm⁻¹ (for energies 1,25 MeV). The protection made of concrete with a filler in the form of lead (leaden fraction) or galena is more compact, but such protection is too much more expensive than usual concretes.

Such X-ray absorbing filler as the baryta BaSO4 allows to solve a problem of choosing width and mass of protection with allowance for its cost. Though the appropriate solution can be found only on the palliative level. The barytes concrete containing as fillers sand and gravel, and the baryta as an X-ray absorbing filler, has density of 3,0 - 3,6 g/cm³, and the linear reduction factor thereof constitutes 0,15 - 0,17 cm⁻¹ (for energies 1,25 MeV). However, the barytes concrete protection total mass for set gamma - quantums energy value remains considerable, which causes serious difficulties while creating protection, especially protection of transport facilities.

The above-stated inconsistency could be overcome, when the iron-manganese concretions are used as an X-ray-absorbing filler, for example, as defined in the patent of Russian Federation № 2029399. But even in this case it is impossible to reduce a total mass of a protective material more than by 20 - 45 %, if compared with the known materials.

However according to the offered invention the correlation of a total mass of segregated poly-dispersed mixture consisting of an X-ray absorbing material particles with the formula set forth above allows to reduce a mass of the included into protective materials known X-ray absorbing fillers in 2 up to 20 times, depending on particular technical conditions and at saving an X-ray and gamma-ray radiation reduction factor.

The technical outcome of the second variant of the invention is obtaining of an X-ray absorbing material with low percentage of a metal-containing X-ray absorbing filler. This effect provides for reduction of width and mass of an X-ray absorbing material as a whole without aggravation of X-ray absorbing properties.

In a third embodiment of an X-ray absorbing material the use of the segregated by intermixing poly-dispersed mixture comprising metallic particles having a size of between 10⁻⁹ up to 10⁻³ m, as a filler, (as was described above) provides for manifestation of qualitatively new effect of the used X-ray absorbing filler, namely, increasing cut of interaction between the X-ray and gamma-ray emission and substance.

The bonding of segregated poly-dispersed mixture consisting of the X-ray absorbing substrate particles to the intermediate substrate promotes obtaining an X-ray absorbing material with even distribution of the heavy X-ray absorbing metal-containing filler inside the matrix having considerably smaller density, than the material of filler.

Allocation of the poly-dispersed mixture comprising metallic particles having a size of between 10⁻⁹ and 10⁻³ m inside the volume of matrix executed of at least one compound solidifying under atmospheric pressure or of the composition on the base of said compound, eliminates (as was described above) violation of the formed at intermixing energetic X-ray absorbing ensembles consisting of the X-ray absorbing element particles poly-dispersed mixture and also promotes self-organizing of energetic X-ray absorbing ensembles.

A textile base and a mineral fiber can be used as an intermediate substrate under the third variant of invention.

The above description of X-ray absorbing material variants confirms the possibility of the invention realization, since the resources known on date of creation of the invention are used. Besides, it is shown that the totality of tags describing an essence of the invention, is sufficient for solution of the set task.

Industrial applicability.

The above stated variants of the invention can be illustrated on the following examples.

Example 1. A filler in the form of segregated by intermixing poly-dispersed mixture made of tungsten particles, is bonded to the matrix surface executed in the form of a twisted lavsan thread. For this purpose a thread is to be put for a period of 10 minutes into the pseudo-liquefied (boiling) (under effect of heavy air stream) stratum of poly-dispersed mixture of the

following faction structure: 20 microns -15 %; 45 microns - 80 %; 500 microns - about 5 %; 1000 microns - 0.01 %.

In these conditions segregation of particles happens due to said particles self-organizing into interdependent power X-ray absorbing ensembles. Meanwhile such particles are attracted to the thread, therefore they are "welded" on its surface. The treated thus thread gains properties providing abnormal reduction of X-radiation.

Data of experiment:

Diameter of a thread - 0,3 mm;

Length of a thread - 3200 mm;

Weight of a thread before plotting mechanical impurity from tungsten - 0,110 g;

Weight of a thread after plotting mechanical impurity from tungsten - 0,160 g;

Solidity of a thread before plotting mechanical impurity from tungsten - 47 H,

the same after plotting mechanical impurity from tungsten - 47 H.

As this has taken place the mass density of ensembles of tungsten particles on the thread surface has constituted $0,0017 \text{ g/cm}^2$, size of the thread - $0,22 \text{ cm}^3$, and density thereof as a whole: $p = 0,7 \text{ g/cm}^3$.

After treating the obtained sample of thread with the stream of quantums with energy of 60 keV and fixing of outcomes on a roentgen film, the densitometry in comparison with the standard leaden plates of different width (stepped weakener of 0,5 mm Pb up to 0,5 mm Pb with step 0,05 mm Pb) has been executed. In outcome it is ascertained that the X-ray absorption of a thread is equivalent to a leaden plate having width of 0,1 mm or 0,075 mm W accordingly, that testifies about abnormally high X-ray absorbing properties of a thread.

Furthermore, according to the formula of the invention

$$\rho = (0.01 - 0.20)\rho_p$$

where ρ m is density of X-ray absorbing material (in our case - a thread) as a whole, while ρ_P - density of X-ray absorbing filler material (in our case - tungsten); we have: ρ m / ρ p = 0,7/19,3=0,036.

The obtained value of the ratio ρ m / ρ p keeps within the range (0,01-0,2) according to the formula of the invention.

Example 2. The segregated poly-dispersed particles of tungsten having a size of between 10⁻⁹ up to 10⁻³ m are bonded to a matrix in the form of a textile material (the thick woolen cloth for

overcoat) having width of 0,4 cm. Segregation and bonding of the tungsten particles to a textile matrix is realized by means of precipitation from hydrosol in conditions of continuous intermixing during the last 15 minutes. Then a sample is to be exsiccated at a room temperature within one day. The subsequent X-ray testing (at quantums energy of 60 keV) has shown that the X-ray protection properties of the obtained sample correspond to the same properties of a leaden slice having width 0,015 cm. This level of protection testifies about abnormally high reduction of X-ray emission stream, since the indicated level of protection at usage of usual non-segregated filler particles material requires bonding to a matrix of 100 % of tungsten by mass (instead of 53 %, as in our case). Indeed, according to the invention and in connection with the considered example the mass of X-ray absorbing filler has constituted 0,116 g, i.e. 53 % of a total mass of a sample, wherein width of a sample made of textile material (the thick woolen cloth for overcoat) has been equal to 0,4 cm, the size of sample has been 1x1 cm² and mass thereof has been 0,216 g. As this has taken place, the density of X-ray absorbing material as a whole has constituted:

$$\rho_{\rm m} = 0.216 / 1 \times 1 \times 0.4 = 0.54 \text{ g/cm}^3$$

and the mass of tungsten of non-segregated particles being equivalent by X-ray-absorbing properties constitutes:

$$0.015 \times 0.75 \times 19.3 = 0.217 g$$

i.e. 100 % of the mass of a textile material sample.

It is obvious therefrom that the relation ρ m / ρ p = 0,54 / 19,3 = 0,0279 corresponds to a declared range.

Example 3. An X-ray absorbing filler in the form of the poly-dispersed particles of tungsten having a size of between 10⁻⁹ and 10⁻³ m, amount = 12 % of mass, is introduced into a matrix in the form of hinge rubber of a brand «Ap - 24» having the following structure: C - 84,73 %; H - 9,12 %; 5 -1,63 %; N - 0,58 %; Zn - 2,27 %; 02 - 1,69 % and a size - 100 cm³. Tungsten particles included into the structure of crude rubber are underwent segregation by intermixing in a mixer during 8 hours. As a result, the self-organizing of particles into the system of power-consuming ensembles is achieved.

After that the crude rubber filled with the X-ray absorbing filler has been underwent vulcanization without effect of pressure. The subsequent X-ray testing (at energy of quantums

- 60 keV) has shown that the X-ray protection properties of the obtained sample of rubber having width 3 mm correspond to the same properties of a leaden slice having width of 0,11 mm. This level of protection testifies about abnormally high reduction of X-ray emission stream, since the indicated level of protection at usage of non-segregated filler particles material requires adding to the matrix of 0,16 g of tungsten, i.e. 34 % by mass (instead of 12 %, as in our case).

Thus, for a considered example (width of a rubber sample -& = 0,3 cm; density - p = $1,56 \text{ g/cm}^3$; a mass of rubber sample having a size 1 x 1 cm constitutes 0,468 g; the filler poly-dispersed particles material total mass, i.e. 12 % of rubber sample mass M=0,056 g) an equivalent mass of X-ray absorbing filler being equal by protective properties to the mass M, is equal m = 0,16 g (34 % of the rubber sample total mass).

It is obvious therefrom that the relation M/m = 0,056 / 0,16 = 0,35 corresponds to the range defined in the formula of invention (0,05 - 0,5), that diminishes the waste of filler, reduces a mass of a protection material as a whole and diminishes the production costs thereof.

Example 4. A filler in the form of super-thin basalt fiber TK-4, on which the segregated by intermixing (in a spherical porcelain attritor) poly-dispersed mixture made of tungsten particles having a size of between 10⁻⁹ and 10⁻³ was fixed, is introduced inside a matrix in the form of epoxy priming of a brand «AP-0010» (Russian Federation Official Standard № 28379-89). A relation of a basalt fiber mass to a mass of tungsten constitutes 1:3. The epoxy priming has been carefully intermixed by a palette-knife with a prepared basalt fiber, thus the relation of a mass of priming to a mass of a fiber has constituted 9:1. After intermixing and obtaining of a homogeneous mass the priming has been spread over a surface of cardboard plates as an even stratum and after solidifying within one day has been tested. The X-ray testing of samples (at energy of quantums - 60 keV) has shown that at a depth of priming layer equal to 2,06 mm, the protective properties thereof are equal to 0,08 mm Pb, that testifies about abnormally high reduction of X-ray emission stream, since the indicated level of protection at usage of non-segregated weighing material particles requires adding to the epoxy matrix 38 % of tungsten by mass (instead of 7,5 %, as in our case).

In a considered example (& = 2,06 mm; p=1,46 g/cm³) the mass of an epoxy priming sample having the size 1x1 cm² constitutes 0,3 g. The total mass of an intermediate substrate with the tungsten particles bonded to the said substrate, constitutes 0,03 g (10 % of a sample mass). As this takes place, the mass of tungsten makes up 3/4 of a mass of the filler, i.e. 0,0225 r, that constitutes 7,5 % of a mass of a sample as a whole.

Furthermore, the mass of tungsten, equal to lead having width of 0,08 mm, constitutes $0.008 \times 0.75 \times 19.3 = 0.1158$ g, which corresponds to 38,6 % of a sample mass.

Example 5. 5 % of mass of the intermediate substrate in the form of crumbled staple fibers (by-products of fulling and worsted industry), to which the poly-dispersed particles of tungsten having a size of between 10⁻⁹ and 10⁻³ m segregated within 20 minutes by intensive intermixing in a pseudo-liquefied layer were bonded, are introduced inside a matrix of dry gypsum. The relation of a mass of staple fibers to a mass of tungsten constitutes 1:3. The prepared thus mixture is carefully intermixed up to obtaining of homogeneous gypsumfilamentary mass. After that water is added, the mass is being carefully intermixed again and samples having sizes of 1x1 cm and width 1 cm are casted of the obtained liquid substance. After drying and solidifying of the samples they are underwent testing (at energy of quantums - 60 keV). The X-ray testing with the subsequent matching with the stepped leaden weakener has shown, that the obtained samples have the protective properties equal to those of a leaden plate having width of 0,04 cm. This level of protection testifies about abnormally high reduction of X-radiation, since the same level of protection can be reached at usage of nonsegregated particles of the filler only at content of tungsten particles - 26,32 % of the mass (instead of 3,75 %, as in our case). For a considered example (width of a gypsum sample -1 cm, density thereof - 1,32 g/cm³) the mass of a sample constitutes 1,32 g. Thus the mass share of tungsten particles in a sample constitutes:

$$1,32 \times 0,05 \times 0,75 = 0,0495 \text{ g},$$

i.e. 3,75 % of the total mass of a sample. At the same time the mass of a tungsten equal to the mass of a leaden plate having width 0,04 cm (by results of the X-ray testing) is equal to $0,04 \times 0,75 \times 19,3 = 0,347 \text{ g}$, that corresponds to 26,32 % of the sample mass.

The above stated examples of particular X-ray absorbing materials embodiment (variants) and the ways of obtaining thereof testify about the industrial applicability of said materials in the indicated area of engineering.

FORMULA OF INVENTION.

1. An X-ray absorbing material comprising a matrix with a fixed X-ray absorbing metal-containing filler in the form of dispersed particles, wherein the said material uses as a filler the segregated by intermixing poly-dispersed mixture containing metallic particles having a size of between 10⁻⁹ and 10⁻³ m while a textile base serves as a matrix; and wherein the particles are bonded to the surface of said textile base, and the density of X-ray absorbing material as a whole, at X-ray absorbing properties of the material being equal to those of the material used for the particles of the X-ray absorbing filler, is defined by the relation:

$$\rho = (0.01 \div 0.20)\rho_p$$

where ρ m is the density of X-ray absorbing material as a whole,

while ρ_P is the density of the material used for the particles of the X-ray absorbing filler.

2. An X-ray absorbing material comprising a matrix with a fixed X-ray absorbing metal-containing filler in the form of dispersed particles, wherein the said material uses as a filler the segregated by intermixing poly-dispersed mixture containing metallic particles having a size of between 10⁻⁹ and 10⁻³ m, wherein the said particles are surrounded by the volume of a matrix made of at least one compound that solidifies under atmospheric pressure or of the composition on the base of said compound, and the total mass of the segregated poly-dispersed mixture consisting of particles of X-ray absorbing filler, is defined by the relation:

$$M = (0.05 \div 0.5) \text{ m}$$

where M is the total mass of the segregated poly-disperse mixture consisting of the X-ray-absorbing filler particles,

while m is the equivalent mass of the X-ray-absorbing filler material equal by its protective properties to the mass M.

3. An X-ray absorbing material comprising a matrix with a fixed X-ray absorbing metal-containing filler in the form of dispersed particles, wherein the said material uses as a filler the segregated by intermixing poly-dispersed mixture containing metallic particles having a size of between 10⁻⁹ and 10⁻³ m, wherein the said particles are bonded to an intermediate substrate surrounded by the volume of a matrix executed of at least one compound that

solidifies under atmospheric pressure or executed of a composition on the base of said compound.

- 4. An X-ray absorbing material as defined in claim 3, wherein a textile base is used as an intermediate substrate.
- 5. An X-ray absorbing material as defined in claim 3, wherein a mineral fiber is used as an intermediate substrate.

Attorney Docket Number: P-0001 ER Nosov et al. * First Named Inventor: **Application No.:** (not yet known)

Filing Date; Group Art Unit; Examiner Name: (not yet known)

UNITED STATES -- PATENT

PCT/RU98/00301

Attorney's Docket No.: P-0001ER

 \square YES \square NO

DECLARATION FOR PATENT APPLICATION

As a below-named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name, I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled X-RAY ABSORBING MATERIAL AND VARIANTS the specification of which

 $(check\ one) \boxtimes is\ attached\ hereto.$ \square was filed on Application Serial No.: _____ (if applicable). and was amended on _____ I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above. I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56(a). I hereby claim foreign priority benefits under Title 35, United States Code, §119 of any foreign application(s) for patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed: Prior Foreign Application(s): **Priority Claimed Date Filed Country** Appln. No. **⊠** YES \square NO 97116386 Russia **September 30, 1997 September 24, 1998** \boxtimes YES **PCT** \square NO

I hereby claim the benefit under Title 35, United States Code, §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose information which is material to patentability as defined in Title 37, Code of Federal Regulations, §1.56(a) which became available between the filing date of the prior application and the national or PCT international filing date of this application:

| Appln. Serial No. | Filing Date | Status: Patented, Pending, Abandoned |
|-------------------|-------------|--------------------------------------|
| | | ☐ Patented ☐ Pending ☐ Abandoned |
| | | ☐ Patented ☐ Pending ☐ Abandoned |

POWER OF ATTORNEY

I hereby appoint the following attorney(s) and/or agent(s) to prosecute the application entitled X-RAY ABSORBING MATERIAL AND VARIANTS and to transact all business in the Patent and Trademark Office connected therewith:

HENRY A. MARZULLO, JR., Reg. No. 20,910;

HOWARD N. ARONSON, Reg. No. 27,302; and

MYRON GREENSPAN, Reg. No. 25,680;

MARVIN FELDMAN, Reg. No. 25,797..

Address all telephone calls to *Myron Greenspan*, at telephone number (914) 723-4300, or to the attorney executing the last document. Address all correspondence to <u>LACKENBACH SIEGEL MARZULLO ARONSON & GREENSPAN</u>, P.C. at Penthouse Suite, One <u>Chase</u> Road, Scarsdale, New York 10583 U.S.A.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the

application or any patent issued thereon.

| Full Name of First or Sole Inventor Igor Stepanovitch NOSOV | Citizenship Russian |
|---|---|
| RESIDENCE Address Street Kv. 20, d. 43, ul. Svedlova | POST OFFICE Address Street (same as residence) |
| City (Zip) Balashiha, Moskovskaya oblast' | City (Zip) |
| State or Country 143900 RUSSIA | State or Country |
| Date 02.25, 2000 | Signature Model |
| Full Name of Second Joint Inventor Vladimir Ivanovich TKATCHENKO | Citizenship Ukrainian |
| RESIDENCE Address Street Kv. 20, d. 4, Pr. K. Marksa | POST OFFICE Address Street (same as residence) |
| City (Zip) 320095 <u>Dnepropetrovsk</u> | City (Zip) |
| | |
| State or Country THE UKRAINE | State or Country |

[■] Additional inventors are being named on separately numbered sheets attached hereto.

New National Phase PCT Application in the U.S. for Nosov et al., Page 3 of 3

| Full Name of Third Joint Inventor <u>Valeriy Anatolievich IVANOV</u> | Citizenship Ukrainian |
|--|--|
| RESIDENCE Address Street Kv. 29, d. 13/15, pr. K. Marksa | POST OFFICE Address - Street (same as residence) |
| City (Zip) 320095 <u>Denpropetrovsk</u> | City (Zip) |
| State or Country THE UKRAINE | State or Country |
| Date 02.25,2000 | Signature Suelency |
| Full Name of Fourth Joint Inventor Valeriy Ivanovich PETCHENKIN | Citizenship Ukrainian |
| RESIDENCE Address Street Kv. 181, d. 80, shosse Zaporozchsky | POST OFFICE Address — Street (same as residence) |
| City (Zip) 320041 Dnepropetrovsk | City (Zip) |
| State or Country THE UKRAINE Date 02.25, 2000 | State or Country |
| Date 02.25, 2000 | Signature Micee |
| Full Name of Fifth Joint Sole Inventor Stanislav Yurivich SOKOLOV | Citizenship Latvian |
| RESIDENCE Address Street Kv. 62, d. 47, ul. Yuglas | POST OFFICE Address Street (same as residence) |
| City (Zip) 1007 Riga | City (Zip) |
| State or Country LATVIA | State or Country |
| Date 02.25, 2000 | Signature Home |

 $[\]square$ Additional inventors are being named on separately numbered sheets attached hereto.